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Les documents fixés à cette attestation sont conformes à la version initialement déposée de la demande de brevet européen spécifiée à la page suivante.

Patentanmeldung Nr.

Patent application No. Demande de brevet nº

03103633.8

PRIORITY DOCUMENT

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For the President of the European Patent Office

Le Président de l'Office européen des brevets p.o.

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Anmeldung Nr:

Application no.:

03103633.8

Demande no:

Anmeldetag:

Date of filing: 01.10.03

Date de dépôt:

Anmelder/Applicant(s)/Demandeur(s):

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Bezeichnung der Erfindung/Title of the invention/Titre de l'invention: (Falls die Bezeichnung der Erfindung nicht angegeben ist, siehe Beschreibung. If no title is shown please refer to the description.
Si aucun titre n'est indiqué se referer à la description.)

Additive Mixtures

In Anspruch genommene Prioriät(en) / Priority(ies) claimed /Priorité(s) revendiquée(s)
Staat/Tag/Aktenzeichen/State/Date/File no./Pays/Date/Numéro de dépôt:

Internationale Patentklassifikation/International Patent Classification/Classification internationale des brevets:

C08K5/00

Am Anmeldetag benannte Vertragstaaten/Contracting states designated at date of filing/Etats contractants désignées lors du dépôt:

AT BE BG CH CY CZ DE DK EE ES FI FR GB GR HU IE IT LU MC NL PT RO SE SI SK TR LI

Additive Mixtures

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The present invention relates to an additive mixture containing a sorbitol derivative, a hydrazine derivative and optionally a lubricant, as well as to an additive mixture containing a sorbitol derivative and a lubricant, to the use of said additive mixtures as clarifying agents for polypropylene homo or copolymers and to polypropylene homo or copolymers containing the described additive mixtures.

The polypropylene homo or copolymers containing the additive mixtures of the present invention exhibit excellent optical properties at considerably reduced side effects (such as discoloration, emission of malodorants, migration or plate-out of low-molecular weight substances etc.) originating e.g. from the additives or any transformation products of the same.

The present invention relates in particular to an additive mixture containing the components (A), (B) and optionally (C), wherein component (A) is at least one compound of the formula (I)

$$(R)_{m}$$
 O
 O
 $(CHOH)_{p}$
 $CH_{2}OH$
 $(R)_{n}$
 (I)

wherein

p is zero or 1;

m and n are independently of one another an integer from zero to 3; and the radicals R are independently of one another C₁-C₈alkyl, C₁-C₈alkoxy, hydroxy, halogen, C₁-C₈alkylthio, C₁-C₈alkylsulfoxy or 2 radicals R form together with 2 adjacent carbon atoms of the unsaturated parent ring a 5- to 7-membered carbocyclic or heterocyclic ring; component (B) is at least one compound of the formula (II)

$$HO \longrightarrow A_{1} \longrightarrow (CH_{2}) \xrightarrow{X} C \longrightarrow NH \longrightarrow NH \longrightarrow C \longrightarrow (CH_{2}) \xrightarrow{A_{1}} OH$$

$$A_{2} \longrightarrow A_{3} \longrightarrow A_{3} \longrightarrow (CH_{2}) \longrightarrow (CH_$$

wherein

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x and y are independently of one another an integer from 2 to 10;

the radicals A_1 , A_2 and A_3 are independently of one another C_1 - C_{10} alkyl, C_5 - C_{12} cycloalkyl unsubstituted or substituted by 1 to 3 C_1 - C_{10} alkyl; phenyl unsubstituted or substituted by 1 to 3 C_1 - C_{10} alkyl; or C_7 - C_{12} phenylalkyl, and

the radicals A2 and A3 are additionally hydrogen;

with the proviso that at least one of the radicals A₁ and A₂ is branched C₃-C₁₀alkyl, C₅-C₁₂cycloalkyl unsubstituted or substituted by 1 to 3 C₁-C₁₀alkyl; phenyl unsubstituted or substituted by 1 to 3 C₁-C₁₀alkyl; or C₇-C₁₂phenylalkyl; and component (C) is a lubricant or a mixture of lubricants.

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The products described as component (A), (B) or (C) are commercially available and can be prepared in analogy to known methods, for example as described in US-A-5,023,354, US-A-5,198,484, US-A-3,660,438 and US-A-3,773,722.

- Examples of alkyl having up to 10 carbon atoms are methyl, ethyl, linear or branched propyl, butyl, pentyl, hexyl, heptyl, octyl, nonyl and decyl. A preferred meaning of R is C₁-C₄alkyl, in particular methyl. A preferred meaning of A₁ and A₂ is branched C₃-C₁₀alkyl, in particular tert-butyl or 1,1-dimethylpropyl. A₁ and A₂ as tert-butyl are especially preferred.
- 20 Preferred meanings of A₃ are hydrogen and methyl.

Examples of C₁-C₈alkoxy are methoxy, ethoxy, propoxy, butoxy, pentyloxy, hexyloxy, heptyloxy and octyloxy. Methoxy is particularly preferred.

25 Examples of C₁-C₈alkylthio are methylthio, ethylthio, propylthio, butylthio, pentylthio, hexylthio and octylthio.

Examples of C_1 - C_8 alkylsulfoxy are methylsulfoxy, ethylsulfoxy, propylsulfoxy, butylsulfoxy, pentylsulfoxy, hexylsulfoxy, heptylsulfoxy and octylsulfoxy.

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Examples of C₅-C₁₂cycloalkyl unsubstituted or substituted by 1 to 3 C₁-C₁₀alkyl are cyclopentyl, cyclohexyl, cyclohexyl, cyclohexyl, cyclohexyl, cyclohexyl.

An example of phenyl substituted by 1 to 3 C₁-C₁₀alkyl is methylphenyl.

Examples of C₇-C₁₂pheniyalkyl are benzyl and 2-phenylprop-2-yl.

An example of the formula (I) wherein 2 radicals R form together with 2 adjacent carbon atoms of the unsaturated parent ring a 5- to 7-membered carbocyclic or heterocyclic ring is

According to a preferred embodiment of the present invention

10 p is 1;

m and n are independently of one another zero, 1 or 2; and the radicals R are independently of one another C₁-C₄alkyl; x and y are independently of one another an integer from 2 to 6; the radicals A₁, A₂ and A₃ are independently of one another C₁-C₅alkyl, cyclohexyl unsubstituted or substituted by one methyl; phenyl unsubstituted or substituted by one methyl; or 2-phenylpropyl, and A₃ is additionally hydrogen.

According to a further preferred embodiment

20 component (A) is at least one compound of the formulae (I-1), (I-2) and (I-3), and

CHOH
$$CH_{2}OH$$

$$CH_{3}C$$

$$CHOH$$

$$CH_{2}OH$$

$$CH_{2}OH$$

$$(I-1)$$

$$H_3C$$
 O
 CH_3
 $CH_$

component (B) is the compound of the formula (II-1).

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$$(H_3C)_3C$$

$$HO \longrightarrow (CH_2)_2 \longrightarrow (CH_2)_2 \longrightarrow (CH_3)_3$$

$$(H_3C)_3C$$

$$(H_3C)_3C$$

$$(CH_3)_3 \longrightarrow (CCH_3)_3$$

$$(II-1)$$

5 The compound of the formula (I-1) is commercially available as IRGACLEAR D (RTM).

The compound of the formula (I-2) is commercially available as IRGACLEAR DM (RTM).

The compound of the formula (I-3) is commercially available as MILLAD 3988 (RTM).

The compound of formula (II-1) is commercially available as IRGANOX MD 1024 (RTM).

- Component (C) is preferably at least one lubricant selected from the group consisting of synthetic or natural waxes and amides of fatty acids. A comprehensive definition and review on waxes is given, for example, in Ullmann's Encyclopedia of Industrial Chemistry, Vol. A-28, VCH Verlagsgesellschaft mbH, D-69451 Weinheim, 1996 (in particular, see pages 104 ff. therein), which is incorporated herewith by reference.
- 20 Preferably suited are synthetic waxes, most preferably fully synthetic waxes of low polarity. Examples are Fischer-Tropsch waxes, high-pressure polyethylene waxes, Ziegler-Natta polyethylene waxes, metallocene polyethylene waxes and Ziegler-Natta polypropylene waxes.
- A most suitable commercially available Fischer-Tropsch wax is for example AdSperse 868 (RTM), available from SASOL, Republic of South Africa.

Most suitable polyolefin waxes have preferably a molecular weight M_w of more than 800 g/mol and less than 20'000 g/mol.

Examples of suitable natural waxes are refined esters of montan wax and decolorized paraffin waxes.

Examples of fatty acid amids are stearamide, erucamide and oleamide which are commercially available as Atmer SA 1750 (RTM), Atmer SA 1753 (RTM), respectively Atmer SA 1756 (RTM), Atmer SA 1758 (RTM) and Atmer SA 1759 (RTM).

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Component (C) is in particular at least one lubricant selected from the group consisting of Fischer-Tropsch wax, high-pressure polyethylene wax, Ziegler-Natta polyethylene wax, metallocene polyethylene wax, Ziegler-Natta polypropylene wax, natural waxes and amides of fatty acids.

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above:

oleamide.

A most particularly preferred embodiment of the present invention relates to an additive mixture wherein component (A) is at least one compound of the formulae (I-1), (I-2) and (I-3) as defined

component (B) is the compound of the formula (II-1) as defined above; and component (C) is at least one lubricant selected from the group consisting of Fischer-Tropsch wax, high-pressure polyethylene wax, Ziegler-Natta polyethylene wax, metallocene polyethylene wax, Ziegler-Natta polypropylene wax and stearamide, erucamide and

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The weight ratio of components (A) to (B) is for example 100/1 to 1/10, more preferably 20/1 to 1/1 and most preferably 5/1 to 2/1.

The weight ratio of components (A) to (C) is for example 1/200 to 100/1, more preferably 1/20 to 10/1 and most preferably 1/10 to 1/1.

Another embodiment of the present invention is a composition containing the components (I) and (II), wherein

component (I) is a polypropylene homopolymer, random copolymer, alternating or segmented copolymer, block copolymer or a blend of polypropylene with another synthetic polymer; and component (II) is the additive mixture described above.

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Component (I) as a polypropylene homopolymer also covers long chain branched polypropylene.

10 The polypropylene, can be prepared by different, and especially by the following, methods:

Catalytic polymerization using a catalyst that normally contains one or more than one metal of groups IVb, Vb, Vlb or VIII of the Periodic Table. These metals usually have one or more than one ligand, typically oxides, halides, alcoholates, esters, ethers, amines, alkyls, alkenyls and/or aryls that may be either π - or σ -coordinated. These metal complexes may be in the free form or fixed on substrates, typically on activated magnesium chloride, titanium(III) chloride, alumina or silicon oxide. These catalysts may be soluble or insoluble in the polymerisation medium. The catalysts can be used by themselves in the polymerisation or further activators may be used, typically metal alkyls, metal hydrides, metal alkyl halides, metal alkyl oxides or metal alkyloxanes, said metals being elements of groups la, Ila and/or Illa of the Periodic Table. The activators may be modified conveniently with further ester, ether, amine or silyl ether groups. These catalyst systems are usually termed Phillips, Standard Oil Indiana, Ziegler (-Natta), TNZ (DuPont), metallocene or single site catalysts (SSC).

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Component (I) is further e.g. a polypropylene random copolymer, alternating or segmented copolymer or block copolymer containing one or more comonomers selected from the group consisting of ethylene, C_4 - C_{20} - α -olefin, vinylcyclohexane, vinylcyclohexene, C_4 - C_{20} alkandiene, C_5 - C_{12} cycloalkandiene and norbornene derivatives; the total amount of propylene and the comonomer(s) being 100 %.

Polypropylene copolymer also covers long chain branched polypropylene copolymer.

Examples of suitable C_4 - $C_{20}\alpha$ -olefins are 1-butene, 1-pentene, 1-hexene, 1-heptene, 1-octene, 1-nonene, 1-decene, 1-undecene, 1-dodecene, 1-tetradecene, 1-hexadecene, 1-octadecene, 1-eicosene and 4-methyl-1-pentene.

5 Examples of suitable C₄-C₂₀alkandienes are hexadiene and octadiene.

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Examples of suitable C_5 - C_{12} cycloalkandienes are cyclopentadiene, cyclohexadiene and cyclooctadiene.

10 Examples of suitable norbornene derivatives are 5-ethylidene-2-norbornene (ENB), dicyclopentadiene (DCP) and methylene-domethylene-hexahydronaphthaline (MEN).

A propylene/ethylene copolymer contains for example 50 to 99.9 %, preferably 80 to 99.9 %, in particular 90 to 99.9 %, by weight of propylene.

A propylene copolymer wherein the comonomer is a C₉-C₂₀α-olefin such as e.g. 1-nonene, 1-decene, 1-undecene, 1-dodecene, 1-tetradecene, 1-hexadecene, 1-octadecene or 1-eicosene; C₉-C₂₀alkandiene, C₉-C₁₂cycloalkandiene or a norbornene derivative such as e.g. 5-ethylidene-2-norbornene (ENB) or methylene-domethylene-hexahydronaphthaline (MEN) contains preferably more than 90 mol %, in particular 90 to 99.9 mol % or 90 to 99 mol %, of propylene.

A propylene copolymer wherein the comonomer is a C_4 - $C_8\alpha$ -olefin such as e.g. 1-butene, 1-pentene, 1-hexene, 1-octene or 4-methyl-1-pentene; vinylcyclohexane, vinylcyclohexane, C_4 - C_8 alkandiene or C_5 - C_8 cycloalkandiene contains preferably more than 80 mol %, in particular 80 to 99.9 mol % or 80 to 99 mol %, of propylene.

Further examples of component (I) are propylene/isobutylene copolymer, propylene/butadiene copolymer, propylene/cycloolefin copolymer, terpolymers of propylene with ethylene and a diene such as hexadiene, dicyclopentadiene or ethylidene-norbornene; propylene/1-olefin copolymers where the 1-olefin is generated in situ; and propylene/carbon monoxide copolymers.

Other examples of component (I) are blends of polypropylene with propylene/ethylene copolymers, propylene/butylene copolymers, polyethylene, e.g. HDPE or LDPE; polybutene, polyisobutylene, poly-4-methylpentene or alternating or random polyalkylene/carbon monoxide copolymers. These blends contain preferably at least 50 % by weight, relative to the weight of the total blend, of polypropylene.

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According to a further preferred embodiment component (I) is a polypropylene copolymer, manufactured by copolymerisation of at least 75 % by weight of propylene with ethylene or another alpha-olefin comonomer, which is selected from linear or branched butene, linear or branched pentene, linear or branched hexene and linear or branched octene.

Component (A) is for example present in the compositions of the invention in an amount of 0.01 to 0.5 % by weight, relative to the weight of component (I), preferably 0.1 to 0.3 % by weight, in particular 0.15 to 0.25 % by weight.

Component (B) is for example present in the compositions of the invention in an amount of 0.01 to 0.5 % by weight, relative to the weight of component (I), preferably 0.01 to 0.3 % by weight, in particular 0.03 to 0.15 % by weight.

Component (C) is for example present in the compositions of the invention in an amount of 0.01 to 5 % by weight, relative to the weight of component (I), preferably 0.1 to 2.0 % by weight, in particular 0.2 to 1.0 % by weight.

25 The compositions of the present invention can be prepared according to the methods well established in the art. Examples are the incorporation of the components of the additive mixture into the polymer resin by powder blending of all components prior to the melt processing; addition of the additive mixture as concentrate or masterbatch to the polymer resin in pellet form; melt blending of the additive mixture in pure or concentrate form, e.g. via continuous feeders or side extruders into the molten polymer resin, etc..

The compositions of the present invention may optionally contain further additives such as those listed below.

1. Antioxidants

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- 1.1. Alkylated monophenols, for example 2,6-di-tert-butyl-4-methylphenol, 2-tert-butyl-4,6-di-methylphenol, 2,6-di-tert-butyl-4-ethylphenol, 2,6-di-tert-butyl-4-n-butylphenol, 2,6-di-tert-butyl-4-methylphenol, 2,6-dioctadecyl-4-methylphenol, 2,4,6-tricyclohexylphenol, 2,6-di-tert-butyl-4-methylphenol, 2,6-dioctadecyl-4-methylphenol, 2,4,6-tricyclohexylphenol, 2,6-di-tert-butyl-4-meth-oxymethylphenol, nonylphenols which are linear or branched in the side chains, for example 2,6-di-nonyl-4-methylphenol, 2,4-dimethyl-6-(1'-methylundec-1'-yl)phenol, 2,4-dimethyl-6-(1'-methyltridec-1'-yl)phenol and mixtures thereof.
- <u>1.2. Alkylthiomethylphenols</u>, for example 2,4-dioctylthiomethyl-6-tert-butylphenol, 2,4-dioctylthiomethyl-6-ethylphenol, 2,6-di-dodecylthiomethyl-4-nonylphenol.
- 1.3. Hydroquinones and alkylated hydroquinones, for example 2,6-di-tert-butyl-4-methoxyphenol, 2,5-di-tert-butylhydroquinone, 2,5-di-tert-amylhydroquinone, 2,6-diphenyl-4-octade-cyloxyphenol, 2,6-di-tert-butylhydroquinone, 2,5-di-tert-butyl-4-hydroxyanisole, 3,5-di-tert-butyl-4-hydroxyphenyl stearate, bis(3,5-di-tert-butyl-4-hydroxyphenyl) adipate.
- <u>1.4. Tocopherols</u>, for example α -tocopherol, β -tocopherol, γ -tocopherol, δ -tocopherol and mixtures thereof (vitamin E).
- 25 <u>1.5. Hydroxylated thiodiphenyl ethers</u>, for example 2,2'-thiobis(6-tert-butyl-4-methylphenol), 2,2'-thiobis(4-octylphenol), 4,4'-thiobis(6-tert-butyl-3-methylphenol), 4,4'-thiobis(6-tert-butyl-2-methylphenol), 4,4'-thiobis(3,6-di-sec-amylphenol), 4,4'-bis(2,6-dimethyl-4-hydroxyphenyl)-disulfide.
- 30 1.6. Alkylidenebisphenols, for example 2,2'-methylenebis(6-tert-butyl-4-methylphenol), 2,2'-methylenebis(6-tert-butyl-4-ethylphenol), 2,2'-methylenebis[4-methyl-6-(α-methylcyclohexyl)-phenol], 2,2'-methylenebis(4-methyl-6-cyclohexylphenol), 2,2'-methylenebis(6-nonyl-4-methylphenol), 2,2'-methylenebis(4,6-di-tert-butyl-phenol), 2,2'-ethylidenebis(6-tert-butyl-4-isobutylphenol), 2,2'-methylenebis[6-(α-methylben-phenol), 2,2'-methylenebis[6-(

zyl)-4-nonylphenol], 2,2'-methylenebis[6-(α , α -dimethylbenzyl)-4-nonylphenol], 4,4'-methylenebis(2,6-di-tert-butylphenol), 4,4'-methylenebis(6-tert-butyl-2-methylphenol), 1,1-bis(5-tert-butyl-4-hydroxy-2-methylphenyl)butane, 2,6-bis(3-tert-butyl-5-methyl-2-hydroxybenzyl)-4-methylphenol, 1,1,3-tris(5-tert-butyl-4-hydroxy-2-methylphenyl)butane, 1,1-bis(5-tert-butyl-4-hydroxy-2-methylphenyl)-3-n-dodecylmercaptobutane, ethylene glycol bis[3,3-bis(3'-tert-butyl-4'-hydroxyphenyl)butyrate], bis(3-tert-butyl-4-hydroxy-5-methyl-phenyl)dicyclopenta-diene, bis[2-(3'-tert-butyl-2'-hydroxy-5'-methylbenzyl)-6-tert-butyl-4-methylphenyl]terephthalate, 1,1-bis-(3,5-dimethyl-2-hydroxyphenyl)butane, 2,2-bis(3,5-di-tert-butyl-4-hydroxyphenyl)propane, 2,2-bis-(5-tert-butyl-4-hydroxy2-methylphenyl)-4-n-dodecylmercaptobutane, 1,1,5,5-tetra(5-tert-butyl-4-hydroxy-2-methylphenyl)pentane.

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- 1.7. O-, N- and S-benzyl compounds, for example 3,5,3',5'-tetra-tert-butyl-4,4'-dihydroxydibenzyl ether, octadecyl-4-hydroxy-3,5-dimethylbenzylmercaptoacetate, tridecyl-4-hydroxy-3,5-di-tert-butyl-4-hydroxybenzyl)amine, bis(4-tert-butyl-3-hydroxy-2,6-dimethylbenzyl)dithioterephthalate, bis(3,5-di-tert-butyl-4-hydroxybenzyl)sulfide, isooctyl-3,5-di-tert-butyl-4-hydroxybenzylmercaptoacetate.
- 1.8. Hydroxybenzylated malonates, for example dioctadecyl-2,2-bis(3,5-di-tert-butyl-2-hydroxybenzyl)malonate, di-octadecyl-2-(3-tert-butyl-4-hydroxy-5-methylbenzyl)malonate, di-octadecylmercaptoethyl-2,2-bis(3,5-di-tert-butyl-4-hydroxybenzyl)malonate, bis[4-(1,1,3,3-te-tramethylbutyl)phenyl]-2,2-bis(3,5-di-tert-butyl-4-hydroxybenzyl)malonate.
- 1.9. Aromatic hydroxybenzyl compounds, for example 1,3,5-tris(3,5-di-tert-butyl-4-hydroxybenzyl)-2,4,6-trimethylbenzene, 1,4-bis(3,5-di-tert-butyl-4-hydroxybenzyl)-2,3,5,6-tetramethylbenzene, 2,4,6-tris(3,5-di-tert-butyl-4-hydroxybenzyl)phenol.
- 1.10. Triazine compounds, for example 2,4-bis(octylmercapto)-6-(3,5-di-tert-butyl-4-hydroxy-anilino)-1,3,5-triazine, 2-octylmercapto-4,6-bis(3,5-di-tert-butyl-4-hydroxyanilino)-1,3,5-triazine, 2-octylmercapto-4,6-bis(3,5-di-tert-butyl-4-hydroxyphenoxy)-1,3,5-triazine, 2,4,6-tris-(3,5-di-tert-butyl-4-hydroxyphenoxy)-1,2,3-triazine, 1,3,5-tris(3,5-di-tert-butyl-4-hydroxyben-zyl)isocyanurate, 1,3,5-tris(4-tert-butyl-3-hydroxy-2,6-dimethylbenzyl)isocyanurate, 2,4,6-tris-(3,5-di-tert-butyl-4-hydroxyphenylethyl)-1,3,5-triazine, 1,3,5-tris(3,5-di-tert-butyl-4-hydroxyphenylpropionyl)-hexahydro-1,3,5-triazine, 1,3,5-tris(3,5-dicyclohexyl-4-hydroxybenzyl)isocyanurate.

- 1.11. Benzylphosphonates, for example dimethyl-2,5-di-tert-butyl-4-hydroxybenzylphosphonate, diethyl-3,5-di-tert-butyl-4-hydroxybenzylphosphonate, dioctadecyl3,5-di-tert-butyl-4-hydroxybenzylphosphonate, dioctadecyl-5-tert-butyl-4-hydroxy-3-methylbenzylphosphonate, the calcium salt of the monoethyl ester of 3,5-di-tert-butyl-4-hydroxybenzylphosphonic acid.
- 1.12. Acylaminophenols, for example 4-hydroxylauranilide, 4-hydroxystearanilide, octyl N-(3,5-di-tert-butyl-4-hydroxyphenyl)carbamate.
- 1.13. Esters of β-(3,5-di-tert-butyl-4-hydroxyphenyl)propionic acid with mono- or polyhydric alcohols, e.g. with methanol, ethanol, n-octanol, i-octanol, octadecanol, 1,6-hexanediol, 1,9-nonanediol, ethylene glycol, 1,2-propanediol, neopentyl glycol, thiodiethylene glycol, diethylene glycol, triethylene glycol, pentaerythritol, tris(hydroxyethyl)isocyanurate, N,N'-bis(hydroxyethyl)oxamide, 3-thiaundecanol, 3-thiapentadecanol, trimethylhexanediol, trimethylol-propane, 4-hydroxymethyl-1-phospha-2,6,7-trioxabicyclo[2.2.2]octane.
 - 1.14. Esters of β-(5-tert-butyl-4-hydroxy-3-methylphenyl)propionic acid with mono- or polyhydric alcohols, e.g. with methanol, ethanol, n-octanol, i-octanol, octadecanol, 1,6-hexanediol, 1,9-nonanediol, ethylene glycol, 1,2-propanediol, neopentyl glycol, thiodiethylene glycol, diethylene glycol, triethylene glycol, pentaerythritol, tris(hydroxyethyl)isocyanurate, N,N'-bis-(hydroxyethyl)oxamide, 3-thiaundecanol, 3-thiapentadecanol, trimethylhexanediol, trimethylolpropane, 4-hydroxymethyl-1-phospha-2,6,7-trioxabicyclo[2.2.2]octane; 3,9-bis[2-{3-(3-tert-butyl-4-hydroxy-5-methylphenyl)propionyloxy}-1,1-dimethylethyl]-2,4,8,10-tetraoxaspiro[5.5]-undecane.

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- 1.15. Esters of β-(3,5-dicyclohexyl-4-hydroxyphenyl)propionic acid with mono- or polyhydric alcohols, e.g. with methanol, ethanol, octanol, octadecanol, 1,6-hexanediol, 1,9-nonanediol, ethylene glycol, 1,2-propanediol, neopentyl glycol, thiodiethylene glycol, diethylene glycol, triethylene glycol, pentaerythritol, tris(hydroxyethyl)isocyanurate, N,N'-bis(hydroxyethyl)oxamide, 3-thiaundecanol, 3-thiapentadecanol, trimethylhexanediol, trimethylolpropane, 4-hydroxymethyl-1-phospha-2,6,7-trioxabicyclo[2.2.2]octane.
- 1.16. Esters of 3,5-di-tert-butyl-4-hydroxyphenyl acetic acid with mono- or polyhydric alcohols, e.g. with methanol, ethanol, octanol, octadecanol, 1,6-hexanediol, 1,9-nonanediol,

ethylene glycol, 1,2-propanediol, neopentyl glycol, thiodiethylene glycol, diethylene glycol, triethylene glycol, pentaerythritol, tris(hydroxyethyl)isocyanurate, N,N'-bis(hydroxyethyl)oxamide, 3-thiaundecanol, 3-thiapentadecanol, trimethylhexanediol, trimethylolpropane, 4-hydroxymethyl-1-phospha-2,6,7-trioxabicyclo[2.2.2]octane.

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1.17. Amides of β-(3,5-di-tert-butyl-4-hydroxyphenyl)propionic acid e.g. N,N'-bis(3,5-di-tert-butyl-4-hydroxyphenylpropionyl)hexamethylenediamide, N,N'-bis(3,5-di-tert-butyl-4-hydroxyphenylpropionyl)trimethylenediamide, N,N'-bis[2-(3-[3,5-di-tert-butyl-4-hydroxyphenyl]-propionyloxy)ethyl]oxamide (Naugard $^{\oplus}$ XL-1, supplied by Uniroyal).

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1.18. Ascorbic acid (vitamin C)

1.19. Aminic antioxidants, for example N,N'-di-isopropyl-p-phenylenediamine, N,N'-di-sec-butyl-p-phenylenediamine, N,N'-bis(1,4-dimethylpentyl)-p-phenylenediamine, N,N'-bis(1-ethyl-3methylpentyl)-p-phenylenediamine, N,N'-bis(1-methylheptyl)-p-phenylenediamine, N,N'-dicyclohexyl-p-phenylenediamine, N,N'-diphenyl-p-phenylenediamine, N,N'-bis(2-naphthyl)-pphenylenediamine, N-isopropyl-N'-phenyl-p-phenylenediamine, N-(1,3-dimethylbutyl)-N'-phenyl-p-phenylenediamine, N-(1-methylheptyl)-N'-phenyl-p-phenylenediamine, N-cyclohexyl-N'phenyl-p-phenylenediamine, 4-(p-toluenesulfamoyl)diphenylamine, N,N'-dimethyl-N,N'-disec-butyl-p-phenylenediamine, diphenylamine, N-allyldiphenylamine, 4-isopropoxydiphenylamine, N-phenyl-1-naphthylamine, N-(4-tert-octylphenyl)-1-naphthylamine, N-phenyl-2-naphthylamine, octylated diphenylamine, for example p,p'-di-tert-octyldiphenylamine, 4-n-butylaminophenol, 4-butyrylaminophenol, 4-nonanoylaminophenol, 4-dodecanoylaminophenol, 4octadecanoylaminophenol, bis(4-methoxyphenyl)amine, 2,6-di-tert-butyl-4-dimethylaminomethylphenol, 2,4'-diaminodiphenylmethane, 4,4'-diaminodiphenylmethane, N,N,N',N'-tetramethyl-4,4'-diaminodiphenylmethane, 1,2-bis[(2-methylphenyl)amino]ethane, 1,2-bis(phenylamino)propane, (o-tolyl)biguanide, bis[4-(1',3'-dimethylbutyl)phenyl]amine, tert-octylated Nphenyl-1-naphthylamine, a mixture of mono- and dialkylated tert-butyl/tert-octyldiphenylamines, a mixture of mono- and dialkylated nonyldiphenylamines, a mixture of mono- and dialkylated dodecyldiphenylamines, a mixture of mono- and dialkylated isopropyl/isohexyldiphenylamines, a mixture of mono- and dialkylated tert-butyldiphenylamines, 2,3-dihydro-3,3-dimethyl-4H-1,4-benzothiazine, phenothiazine, a mixture of mono- and dialkylated tertbutyl/tert-octylphenothiazines, a mixture of mono- and dialkylated tert-octylphenothiazines, N-allylphenothiazine. N,N,N',N'-tetraphenyl-1,4-diaminobut-2-ene, N,N-bis(2,2,6,6-tetramethylpiperid-4-yl-hexamethylenediamine, bis(2,2,6,6-tetramethylpiperid-4-yl)sebacate, 2,2,6,6-tetramethylpiperidin-4-one, 2,2,6,6-tetramethylpiperidin-4-ol.

2. UV absorbers and light stabilisers

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- 2.1. 2-(2'-Hydroxyphenyl)benzotriazoles, for example 2-(2'-hydroxy-5'-methylphenyl)benzotriazole, 2-(3',5'-di-tert-butyl-2'-hydroxyphenyl)benzotriazole, 2-(5'-tert-butyl-2'-hydroxyphenyl)benzotriazole, 3-(5'-tert-butyl-2'-hydroxyphenyl)benzotriazole, 3-(5'-tert-butyl-2'-h nyl)benzotriazole, 2-(2'-hydroxy-5'-(1,1,3,3-tetramethylbutyl)phenyl)benzotriazole, 2-(3',5'-ditert-butyl-2'-hydroxyphenyl)-5-chlorobenzotriazole, 2-(3'-tert-butyl-2'-hydroxy-5'-methylphenyl)-5-chlorobenzotriazole, 2-(3'-sec-butyl-5'-tert-butyl-2'-hydroxyphenyl)benzotriazole, 2-(2'hydroxy-4'-octyloxyphenyl)benzotriazole, 2-(3',5'-di-tert-amyl-2'-hydroxyphenyl)benzotriazole, 2-(3',5'-bis(α,α-dimethylbenzyl)-2'-hydroxyphenyl)benzotriazole, 2-(3'-tert-butyl-2'-hydroxy-5'-(2-octyloxycarbonylethyl)phenyl)-5-chlorobenzotriazole, 2-(3'-tert-butyl-5'-[2-(2-ethylhexyloxy)carbonylethyl]-2'-hydroxyphenyl)-5-chlorobenzotriazole, 2-(3'-tert-butyl-2'-hydroxy-5'-(2methoxycarbonylethyl)phenyl)-5-chlorobenzotriazole, 2-(3'-tert-butyl-2'-hydroxy-5'-(2-methoxycarbonylethyl)phenyl)benzotriazole, 2-(3'-tert-butyl-2'-hydroxy-5'-(2-octyloxycarbonyl-2-(3'-tert-butyi-5'-[2-(2-ethylhexyloxy)carbonylethyl]-2'-hydroxyethyl)phenyl)benzotriazole, phenyl)benzotriazole, 2-(3'-dodecyl-2'-hydroxy-5'-methylphenyl)benzotriazole, 2-(3'-tert-butyl-2'-hydroxy-5'-(2-isooctyloxycarbonylethyl)phenylbenzotriazole, 2,2'-methylenebis[4-(1,1,3,3tetramethylbutyl)-6-benzotriazole-2-ylphenol]; the transesterification product of 2-[3'-tert-butyl-5'-(2-methoxycarbonylethyl)-2'-hydroxyphenyl]-2H-benzotriazole with polyethylene glycol $[R-CH_2CH_2-COO-CH_2CH_2]_2$, where R = 3'-tert-butyl-4'-hydroxy-5'-2H-benzotri-2-[2'-hydroxy-3'- $(\alpha,\alpha$ -dimethylbenzyl)-5'-(1,1,3,3-tetramethylbutyl)phenyl]azol-2-ylphenyl, benzotriazole; 2-[2'-hydroxy-3'-(1,1,3,3-tetramethylbutyl)-5'-(α,α-dimethylbenzyl)phenyl]benzotriazole.
- <u>2.2. 2-Hydroxybenzophenones</u>, for example the 4-hydroxy, 4-methoxy, 4-octyloxy, 4-decyloxy, 4-dodecyloxy, 4-benzyloxy, 4,2',4'-trihydroxy and 2'-hydroxy-4,4'-dimethoxy derivatives.
- 30 <u>2.3. Esters of substituted and unsubstituted benzoic acids</u>, for example 4-tert-butylphenyl salicylate, phenyl salicylate, octylphenyl salicylate, dibenzoyl resorcinol, bis(4-tert-butylbenzoyl)resorcinol, benzoyl resorcinol, 2,4-di-tert-butylphenyl 3,5-di-tert-butyl-4-hydroxybenzo-

ate, hexadecyl 3,5-di-tert-butyl-4-hydroxybenzoate, octadecyl 3,5-di-tert-butyl-4-hydroxybenzoate, 2-methyl-4,6-di-tert-butylphenyl 3,5-di-tert-butyl-4-hydroxybenzoate.

2.4. Acrylates, for example ethyl α-cyano- β , β -diphenylacrylate, isooctyl α-cyano- β , β -diphenylacrylate, methyl α-carbomethoxycinnamate, methyl α-cyano- β -methyl-p-methoxycinnamate, butyl α-cyano- β -methyl-p-methoxycinnamate, methyl α-carbomethoxy-p-methoxycinnamate and N-(β -carbomethoxy- β -cyanovinyl)-2-methylindoline.

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- 2.5. Nickel compounds, for example nickel complexes of 2,2'-thiobis[4-(1,1,3,3-tetramethyl-butyl)phenol], such as the 1:1 or 1:2 complex, with or without additional ligands such as n-butylamine, triethanolamine or N-cyclohexyldiethanolamine, nickel dibutyldithiocarbamate, nickel salts of the monoalkyl esters, e.g. the methyl or ethyl ester, of 4-hydroxy-3,5-di-tert-butylbenzylphosphonic acid, nickel complexes of ketoximes, e.g. of 2-hydroxy-4-methylphenylundecylketoxime, nickel complexes of 1-phenyl-4-lauroyl-5-hydroxypyrazole, with or without additional ligands.
 - 2.6. Sterically hindered amines, for example bis(2,2,6,6-tetramethyl-4-piperidyl)sebacate, bis(2,2,6,6-tetramethyl-4-piperidyl)succinate, bis(1,2,2,6,6-pentamethyl-4-piperidyl)sebacate, bis(1-octyloxy-2,2,6,6-tetramethyl-4-piperidyl)sebacate. bis(1,2,2,6,6-pentamethyl-4-piperidyl) n-butyl-3,5-di-tert-butyl-4-hydroxybenzylmalonate, the condensate of 1-(2-hydroxyethyl)-2,2,6,6-tetramethyl-4-hydroxypiperidine and succinic acid, linear or cyclic condensates of N,N'-bis(2,2,6,6-tetramethyl-4-piperidyl)hexamethylenediamine and 4-tert-octylamino-2,6-dichloro-1,3,5-triazine, tris(2,2,6,6-tetramethyl-4-piperidyl)nitrilotriacetate, tetrakis(2,2,6,6-tetramethyl-4-piperidyl)-1,2,3,4-butanetetracarboxylate, 1,1'-(1,2-ethanediyl)-bis(3,3,5,5-tetramethylpiperazinone), 4-benzoyl-2,2,6,6-tetramethylpiperidine, 4-stearyloxy-2,2,6,6-tetramethylpiperidine, bis(1,2,2,6,6-pentamethylpiperidyl)-2-n-butyl-2-(2-hydroxy-3,5-di-tert-butylbenzyl)malonate, 3-n-octyl-7,7,9,9-tetramethyl-1,3,8-triazaspiro[4.5]decane-2,4-dione, bis(1-octyloxy-2,2,6,6-tetramethylpiperidyl)sebacate, bis(1-octyloxy-2,2,6,6-tetramethylpiperidyl)succinate, linear or cyclic condensates of N,N'-bis(2,2,6,6-tetramethyl-4-piperidyl)hexamethylenediamine and 4-morpholino-2,6-dichloro-1,3,5-triazine, the condensate of 2-chloro-4,6-bis(4-nbutylamino-2,2,6,6-tetramethylpiperidyl)-1,3,5-triazine and 1,2-bis(3-aminopropylamino)ethane, the condensate of 2-chloro-4,6-di-(4-n-butylamino-1,2,2,6,6-pentamethylpiperidyl)-1.3.5-triazine and 1,2-bis(3-aminopropylamino)ethane, 8-acetyl-3-dodecyl-7,7,9,9-tetramethyl-1,3,8-triazaspiro[4.5]decane-2,4-dione, 3-dodecyl-1-(2,2,6,6-tetramethyl-4-piperidyl)pyr-

rolidine-2,5-dione, 3-dodecyl-1-(1,2,2,6,6-pentamethyl-4-piperidyl)pyrrolidine-2,5-dione, mixture of 4-hexadecyloxy- and 4-stearyloxy-2,2,6,6-tetramethylpiperidine, a condensate of N,N'-bis(2,2,6,6-tetramethyl-4-piperidyl)hexamethylenediamine and 4-cyclohexylamino-2,6dichloro-1,3,5-triazine, a condensate of 1,2-bis(3-aminopropylamino)ethane and 2,4,6-trichloro-1,3,5-triazine as well as 4-butylamino-2,2,6,6-tetramethylpiperidine (CAS Reg. No. [136504-96-6]); a condensate of 1,6-hexanediamine and 2,4,6-trichloro-1,3,5-triazine as well as N,N-dibutylamine and 4-butylamino-2,2,6,6-tetramethylpiperidine (CAS Reg. No. N-(2,2,6,6-tetramethyl-4-piperidyl)-n-dodecylsuccinimide, N-(1,2,2,6,6-[192268-64-7]); pentamethyl-4-piperidyl)-n-dodecylsuccinimide, 2-undecyl-7,7,9,9-tetramethyl-1-oxa-3,8-diaza-4-oxo-spiro[4,5]decane, a reaction product of 7,7,9,9-tetramethyl-2-cycloundecyl-1-oxa-1,1-bis(1,2,2,6,6-pentamethyl-4-3,8-diaza-4-oxospiro-[4,5]decane and epichlorohydrin, piperidyloxycarbonyl)-2-(4-methoxyphenyl)ethene, N,N'-bis-formyl-N,N'-bis(2,2,6,6-tetramethyl-4-piperidyl)hexamethylenediamine, a diester of 4-methoxymethylenemalonic acid with 1,2,2,6,6-pentamethyl-4-hydroxypiperidine, poly[methylpropyl-3-oxy-4-(2,2,6,6-tetramethyl-4-15 piperidyl)]siloxane, a reaction product of maleic acid anhydride-α-olefin copolymer with 2,2,6,6-tetramethyl-4-aminopiperidine or 1,2,2,6,6-pentamethyl-4-aminopiperidine.

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2.7. Oxamides, for example 4,4'-dioctyloxyoxanilide, 2,2'-diethoxyoxanilide, 2,2'-dioctyloxy-5,5'-di-tert-butoxanilide, 2,2'-didodecyloxy-5,5'-di-tert-butoxanilide, 2-ethoxy-2'-ethyloxanilide, N,N'-bis(3-dimethylaminopropyl)oxamide, 2-ethoxy-5-tert-butyl-2'-ethoxanilide and its mixture with 2-ethoxy-2'-ethyl-5,4'-di-tert-butoxanilide, mixtures of o- and p-methoxy-disubstituted oxanilides and mixtures of o- and p-ethoxy-disubstituted oxanilides.

2.8. 2-(2-Hydroxyphenyl)-1,3,5-triazines, for example 2,4,6-tris(2-hydroxy-4-octyloxyphenyl)-1,3,5-triazine, 2-(2-hydroxy-4-octyloxyphenyl)-4,6-bis(2,4-dimethylphenyl)-1,3,5-triazine, 2-(2,4-dihydroxyphenyl)-4,6-bis(2,4-dimethylphenyl)-1,3,5-triazine, 2,4-bis(2-hydroxy-4-propyloxyphenyl)-6-(2,4-dimethylphenyl)-1,3,5-triazine, 2-(2-hydroxy-4-octyloxyphenyl)-4,6-bis(4methylphenyl)-1,3,5-triazine, 2-(2-hydroxy-4-dodecyloxyphenyl)-4,6-bis(2,4-dimethylphenyl)-1,3,5-triazine, 2-(2-hydroxy-4-tridecyloxyphenyl)-4,6-bis(2,4-dimethylphenyl)-1,3,5-triazine, 2-[2-hydroxy-4-(2-hydroxy-3-butyloxypropoxy)phenyl]-4,6-bis(2,4-dimethyl)-1,3,5-triazine, 2-[2hydroxy-4-(2-hydroxy-3-octyloxypropyloxy)phenyl]-4,6-bis(2,4-dimethyl)-1,3,5-triazine, (dodecyloxy/tridecyloxy-2-hydroxypropoxy)-2-hydroxyphenyl]-4,6-bis(2,4-dimethylphenyl)-1,3,5-triazine, 2-[2-hydroxy-4-(2-hydroxy-3-dodecyloxypropoxy)phenyl]-4,6-bis(2,4-dimethylphenyl)-1,3,5-triazine, 2-(2-hydroxy-4-hexyloxy)phenyl-4,6-diphenyl-1,3,5-triazine, 2-(2-hydroxy-4-methoxyphenyl)-4,6-diphenyl-1,3,5-triazine, 2,4,6-tris[2-hydroxy-4-(3-butoxy-2-hydroxypropoxy)phenyl]-1,3,5-triazine, 2-(2-hydroxyphenyl)-4-(4-methoxyphenyl)-6-phenyl-1,3,5-triazine, 2-{2-hydroxy-4-[3-(2-ethylhexyl-1-oxy)-2-hydroxypropyloxy]phenyl}-4,6-bis(2,4-dimethylphenyl)-1,3,5-triazine.

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- 3. Metal deactivators, for example N,N'-diphenyloxamide, N-salicylal-N'-salicyloyl hydrazine, N,N'-bis(salicyloyl)hydrazine, 3-salicyloylamino-1,2,4-triazole, bis(benzylidene)oxalyl dihydrazide, oxanilide, isophthaloyl dihydrazide, sebacoyl bisphenylhydrazide, N,N'-diacetyladipoyl dihydrazide, N,N'-bis(salicyloyl)oxalyl dihydrazide, N,N'-bis(salicyloyl)thiopropionyl dihydrazide.
- 4. Phosphites and phosphonites, for example triphenyl phosphite, diphenylalkyl phosphites, phenyldialkyl phosphites, tris(nonylphenyl) phosphite, trilauryl phosphite, trioctadecyl phosphite, distearylpentaerythritol diphosphite, tris(2,4-di-tert-butylphenyl) phosphite, diisodecyl pentaerythritol diphosphite, bis(2,4-di-tert-butylphenyl)pentaerythritol diphosphite, bis(2,4-di-tert-butyl-4-methylphenyl)pentaerythritol diphosphite, diisodecyloxypentaerythritol diphosphite, bis(2,4-di-tert-butyl-6-methylphenyl)pentaerythritol diphosphite, bis(2,4-6-tris(tert-butylphenyl)pentaerythritol diphosphite, tristearyl sorbitol triphosphite, tetrakis(2,4-di-tert-butylphenyl) 4,4'-biphenylene diphosphonite, 6-isooctyloxy-2,4,8,10-tetra-tert-butyl-12H-dibenz[d,g]-1,3,2-dioxaphosphocin, bis(2,4-di-tert-butyl-6-methylphenyl)methyl phosphite, bis(2,4-di-tert-butyl-6-methylphenyl)ethyl phosphite, 6-fluoro-2,4,8,10-tetra-tert-butyl-12-methyl-dibenz[d,g]-1,3,2-dioxaphosphocin, 2,2',2"-nitrilo-[triethyltris(3,3',5,5'-tetra-tert-butyl-1,1'-biphenyl-2,2'-diyl)phosphite], 2-ethylhexyl(3,3',5,5'-tetra-tert-butyl-1,1'-biphenyl-2,2'-diyl)phosphite, 5-butyl-5-ethyl-2-(2,4,6-tri-tert-butylphenoxy)-1,3,2-dioxaphosphirane.

The following phosphites are especially preferred:

Tris(2,4-di-tert-butylphenyl) phosphite (Irgafos®168, Ciba-Geigy), tris(nonylphenyl) phosphite,

(CH₃)₃C
$$C(CH_3)_3$$
 $C(CH_3)_3$ $C(CH$

$$(CH_3)_3C$$
 $C(CH_3)_3$
 C
 $C(CH_3)_3$
 C
 $C(CH_3)_3$
 C
 $C(CH_3)_3$
 C
 $C(CH_3)_3$

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$$(CH_3)_3C - (CH_3)_3 - (CH_3)_3 (D)$$

$$H_3C$$
 $C(CH_3)_3$
 $C(CH_3)_3$

(F)
$$H_{37}C_{18} O - P_{O} O P - O - C_{18}H_{37}$$

$$H_{3}C - C - CH_{3}$$

$$H_{3}C - C - CH_{3}$$

$$H_{3}C - C - CH_{3}$$

$$CH_{3} - C - CH_{3}$$

$$CH_{3} - C - CH_{3}$$

<u>5. Hydroxylamines</u>, for example N,N-dibenzylhydroxylamine, N,N-diethylhydroxylamine, N,N-dioctylhydroxylamine, N,N-diletradecylhydroxylamine, N,N-dihexadecylhydroxylamine, N,N-dioctadecylhydroxylamine, N-hexadecyl-N-octadecylhydroxylamine, N,N-dialkylhydroxylamine derived from hydrogenated tallow amine.

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- 6. Nitrones, for example N-benzyl-alpha-phenylnitrone, N-ethyl-alpha-methylnitrone, N-octyl-alpha-heptylnitrone, N-lauryl-alpha-undecylnitrone, N-tetradecyl-alpha-tridecylnitrone, N-hexadecyl-alpha-heptadecylnitrone, N-octadecyl-alpha-heptadecylnitrone, N-hexadecyl-alpha-heptadecylnitrone, N-octadecyl-alpha-pentadecylnitrone, N-heptadecyl-alpha-heptadecylnitrone, N-octadecyl-alpha-hexadecylnitrone, nitrone derived from N,N-dialkylhydroxyl-amine derived from hydrogenated tallow amine.
- 7. Thiosynergists, for example dilauryl thiodipropionate or distearyl thiodipropionate.
- 8. Peroxide scavengers, for example esters of β -thiodipropionic acid, for example the lauryl, stearyl, myristyl or tridecyl esters, mercaptobenzimidazole or the zinc salt of 2-mercaptobenzimidazole, zinc dibutyldithiocarbamate, dioctadecyl disulfide, pentaerythritol tetrakis(β -dodecylmercapto)propionate.
- 9. Polyamide stabilisers, for example copper salts in combination with iodides and/or phosphorus compounds and salts of divalent manganese.
- 10. Basic co-stabilisers, for example melamine, polyvinylpyrrolidone, dicyandiamide, triallyl cyanurate, urea derivatives, hydrazine derivatives, amines, polyamides, polyurethanes, alkali metal salts and alkaline earth metal salts of higher fatty acids, for example calcium stearate, zinc stearate, magnesium behenate, magnesium stearate, sodium ricinoleate and potassium palmitate, antimony pyrocatecholate or zinc pyrocatecholate.
- 30 11. Conventional nucleating agents, for example inorganic substances, such as talcum, metal oxides, such as titanium dioxide or magnesium oxide, phosphates, carbonates or sulfates of, preferably, alkaline earth metals; organic compounds, such as mono- or polycarboxylic acids and the salts thereof, e.g. 4-tert-butylbenzoic acid, adipic acid,

diphenylacetic acid, sodium succinate or sodium benzoate; polymeric compounds, such as ionic copolymers (ionomers).

- 12. Other additives, for example plasticisers, lubricants, rheology additives, catalysts, flow-control agents, optical brighteners, flame retarding agents, antistatic agents and blowing agents.
 - 13. Benzofuranones and indolinones, for example those disclosed in US-A-4,325,863; US-A-4,338,244; US-A-5,175,312; US-A-5,216,052; US-A-5,252,643; DE-A-4316611;
- DE-A-4316622; DE-A-4316876; EP-A-0589839 or EP-A-0591102 or 3-[4-(2-acetoxyethoxy)-phenyl]-5,7-di-tert-butylbenzofuran-2-one, 5,7-di-tert-butyl-3-[4-(2-stearoyloxyethoxy)phenyl]-benzofuran-2-one, 3,3'-bis[5,7-di-tert-butyl-3-(4-[2-hydroxyethoxy]phenyl)benzofuran-2-one], 5,7-di-tert-butyl-3-(4-ethoxyphenyl)benzofuran-2-one, 3-(4-acetoxy-3,5-dimethylphenyl)-5,7-di-tert-butylbenzofuran-2-one, 3-(3,5-dimethyl-4-pivaloyloxyphenyl)-5,7-di-tert-butylbenzofuran-2-one, 3-(3,4-dimethylphenyl)-5,7-di-tert-butylbenzofuran-2-one, 3-(2,3-dimethylphenyl)-5,7-di-tert-butylbenzofuran-2-one.

The weight ratio of the above described component (A) to the total of the conventional additives is preferably 250/1 to 1/2500, more preferably 100/1 to 1/100 and most preferably 10/1 to 1/10.

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Further embodiments of the present invention are
the use of the additive mixture described above as clarifying agent for a polypropylene
homopolymer, random copolymer, alternating or segmented copolymer, block copolymer or a
blend of polypropylene with another synthetic polymer; and
a method for clarifying a polypropylene homopolymer, random copolymer, alternating or
segmented copolymer, block copolymer or a blend of polypropylene with another synthetic
polymer, which comprises incorporating therein an additive mixture as described above.

30 The present invention also relates to an additive mixture containing the components (A') and (C'), wherein component (A') is at least one of the compounds of the formulae (I-1) and (I-2); and

CHOH
$$CH_{2}OH$$

$$CH_{3}C$$

$$CHOH$$

$$CH_{2}OH$$

$$CH_{2}OH$$

$$(I-1)$$

$$CH_{2}OH$$

$$(I-2)$$

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component (C') is at least one lubricant selected from the group consisting of synthetic waxes, preferably polyolefin waxes, in particular Fischer-Tropsch wax, high-pressure polyethylene wax, Ziegler-Natta polyethylene wax, metallocene polyethylene wax or Ziegler-Natta polypropylene wax; and stearamide, erucamide and oleamide; as well as to

an additive mixture containing the components (A") and (C") wherein component (A") is the compound of the formula (I-3); and

$$H_3C$$
 O
 CH_3
 $CHOH$
 CH_2OH
 CH_3
 C

component (C") is selected from the group consisting of stearamide, erucamide and oleamide.

The particular uses and applications described herein are also applicable in appropriate manner for the latter two additive mixtures.

The weight ratios and concentrations given for components (A), (C) and (I) can be applied in appropriate manner for components (A'), (A"), (C'), (C"), (I') and (I") so that component (A) corresponds to component (A') or (A"), component (C) corresponds to component (C') or (C") and component (I) corresponds to component (I') or (I").

The present invention further relates to a composition containing the components (I"'), (A"') and (C"') wherein

component (I"") is a polypropylene homopolymer or a polypropylene copolymer, manufactured by copolymerisation of at least 75 % by weight of propylene with ethylene or another alpha-olefin comonomer, which is selected from linear or branched butene, linear or branched pentene, linear or branched hexene and linear or branched octene;

5 component (A"") is the compound of the formula (I-3); and

$$H_3C$$
 O
 $CHOH$
 CH_3
 $CH_$

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component (C"") is selected from the group consisting of Fischer-Tropsch wax, high-pressure polyethylene wax, Ziegler-Natta polyethylene wax, metallocene polyethylene wax, Ziegler-Natta polypropylene wax and stearamide, erucamide and oleamide.

The weight ratios and concentrations given for components (A), (C) and (I) can be applied in appropriate manner for components (A""), (C""), and (I""). The applications and uses described herein also apply to the latter composition.

The polymer resin compositions according to the present invention can be advantageously used for the preparation of various shaped articles. Examples are:

I-1) Floating devices, marine applications, pontoons, buoys, plastic lumber for decks, piers, boats, kayaks, oars, and beach reinforcements.

I-2) Automotive applications, in particular bumpers, dashboards, battery, rear and front linings, moldings parts under the hood, hat shelf, trunk linings, interior linings, air bag covers, electronic moldings for fittings (lights), panes for dashboards, headlamp glass, instrument panel, exterior linings, upholstery, automotive lights, head lights, parking lights, rear lights, stop lights, interior and exterior trims; door panels; gas tank; glazing front side; rear windows; seat backing, exterior panels, wire insulation, profile extrusion for sealing, cladding, pillar covers, chassis parts, exhaust systems, fuel filter / filler, fuel pumps, fuel tank, body side mouldings, convertible tops, exterior mirrors, exterior trim, fasteners / fixings, front end module, glass, hinges, lock systems, luggage / roof racks, pressed/stamped parts, seals, side impact protection, sound deadener / insulator and sunroof.

- I-3) Road traffic devices, in particular sign postings, posts for road marking, car accessories, warning triangles, medical cases, helmets, tires.
- 5 I-4) Devices for plane, railway, motor car (car, motorbike) including furnishings.
 - I-5) Devices for space applications, in particular rockets and satellites, e.g. reentry shields.
- I-6) Devices for architecture and design, mining applications, acoustic quietized systems,street refuges, and shelters.
 - II-1) Appliances, cases and coverings in general and electric/electronic devices (personal computer, telephone, portable phone, printer, television-sets, audio and video devices), flower pots, satellite TV bowl, and panel devices.
 - II-2) Jacketing for other materials such as steel or textiles.

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- II-3) Devices for the electronic industry, in particular insulation for plugs, especially computer plugs, cases for electric and electronic parts, printed boards, and materials for electronic data storage such as chips, check cards or credit cards.
- II-4) Electric appliances, in particular washing machines, tumblers, ovens (microwave oven), dish-washers, mixers, and irons.
- 25 II-5) Covers for lights (e.g. street-lights, lamp-shades).
 - II-6) Applications in wire and cable (semi-conductor, insulation and cable-jacketing).
- II-7) Foils for condensers, refrigerators, heating devices, air conditioners, encapsulating of
 electronics, semi-conductors, coffee machines, and vacuum cleaners.
 - III-1) Technical articles such as cogwheel (gear), slide fittings, spacers, screws, bolts, handles, and knobs.

III-2) Rotor blades, ventilators and windmill vanes, solar devices, swimming pools, swimming pool covers, pool liners, pond liners, closets, wardrobes, dividing walls, slat walls, folding walls, roofs, shutters (e.g. roller shutters), fittings, connections between pipes, sleeves, and conveyor belts.

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- III-3) Sanitary articles, in particular shower cubicles, lavatory seats, covers, and sinks.
- III-4) Hygienic articles, in particular diapers (babies, adult incontinence), feminine hygiene articles, shower curtains, brushes, mats, tubs, mobile toilets, tooth brushes, and bed pans.

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- III-5) Pipes (cross-linked or not) for water, waste water and chemicals, pipes for wire and cable protection, pipes for gas, oil and sewage, guttering, down pipes, and drainage systems.
- 15 III-6) Profiles of any geometry (window panes) and siding.
 - III-7) Glass substitutes, in particular extruded plates, glazing for buildings (monolithic, twin or multiwall), aircraft, schools, extruded sheets, window film for architectural glazing, train, transportation, sanitary articles, and greenhouse.

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III-8) Plates (walls, cutting board), extrusion-coating (photographic paper, tetrapack and pipe coating), silos, wood substitute, plastic lumber, wood composites, walls, surfaces, furniture, decorative foil, floor coverings (interior and exterior applications), flooring, duck boards, and tiles.

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- III-9) Intake and outlet manifolds.
- III-10) Cement-, concrete-, composite-applications and covers, siding and cladding, hand rails, banisters, kitchen work tops, roofing, roofing sheets, tiles, and tarpaulins.

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IV-1) Plates (walls and cutting board), trays, artificial grass, astroturf, artificial covering for stadium rings (athletics), artificial floor for stadium rings (athletics), and tapes.

IV-2) Woven fabrics continuous and staple, fibers (carpets / hygienic articles / geotextiles / monofilaments; filters; wipes / curtains (shades) / medical applications), bulk fibers (applications such as gown / protection clothes), nets, ropes, cables, strings, cords, threads, safety seat-belts, clothes, underwear, gloves; boots; rubber boots, intimate apparel, garments, swimwear, sportswear, umbrellas (parasol, sunshade), parachutes, paraglides, sails, "balloon-silk", camping articles, tents, airbeds, sun beds, bulk bags, and bags.

IV-3) Membranes, insulation, covers and seals for roofs, tunnels, dumps, ponds, dumps, walls roofing membranes, geomembranes, swimming pools, curtains (shades) / sun-shields, awnings, canopies, wallpaper, food packing and wrapping (flexible and solid), medical packaging (flexible & solid), airbags/safety belts, arm- and head rests, carpets, centre console, dashboard, cockpits, door, overhead console module, door trim, headliners, interior lighting, interior mirrors, parcel shelf, rear luggage cover, seats, steering column, steering wheel, textiles, and trunk trim.

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- V) Films (packaging, dump, laminating, agriculture and horticulture, greenhouse, mulch, tunnel, silage), bale wrap, swimming pools, waste bags, wallpaper, stretch film, raffia, desalination film, batteries, and connectors.
- 20 VI-1) Food packing and wrapping (flexible and solid), BOPP, BOPET, bottles.
 - VI-2) Storage systems such as boxes (crates), luggage, chest, household boxes, pallets, shelves, tracks, screw boxes, packs, and cans.
- VI-3) Cartridges, syringes, medical applications, containers for any transportation, waste baskets and waste bins, waste bags, bins, dust bins, bin liners, wheely bins, container in general, tanks for water / used water / chemistry / gas / oil / gasoline / diesel; tank liners, boxes, crates, battery cases, troughs, medical devices such as piston, ophthalmic applications, diagnostic devices, and packing for pharmaceuticals blister.

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VII-1) Extrusion coating (photo paper, tetrapack, pipe coating), household articles of any kind (e.g. appliances, thermos bottle / clothes hanger), fastening systems such as plugs, wire and cable clamps, zippers, closures, locks, and snap-closures.

- VII-2) Support devices, articles for the leisure time such as sports and fitness devices, gymnastics mats, ski-boots, inline-skates, skis, big foot, athletic surfaces (e.g. tennis grounds); screw tops, tops and stoppers for bottles, and cans.
- VII-3) Furniture in general, foamed articles (cushions, impact absorbers), foams, sponges, dish clothes, mats, garden chairs, stadium seats, tables, couches, toys, building kits (boards / figures / balls), playhouses, slides, and play vehicles.
 - VII-4) Materials for optical and magnetic data storage.

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- VII-5) Kitchen ware (eating, drinking, cooking, storing).
- VII-6) Boxes for CD's, cassettes and video tapes; DVD electronic articles, office supplies of any kind (ball-point pens, stamps and ink-pads, mouse, shelves, tracks), bottles of any volume and content (drinks, detergents, cosmetics including perfumes), and adhesive tapes.
 - VII-7) Footwear (shoes / shoe-soles), insoles, spats, adhesives, structural adhesives, food boxes (fruit, vegetables, meat, fish), synthetic paper, labels for bottles, couches, artificial joints (human), printing plates (flexographic), printed circuit boards, and display technologies.
- VII-8) Devices of filled polymers (talc, chalk, china clay (kaolin), wollastonite, pigments, carbon black, TiO₂, mica, nanocomposites, dolomite, silicates, glass, asbestos).
- Thus, a preferred embodiment of the present invention relates to shaped articles, in particular containers or boxes for food packaging and storage, containers or boxes for non-food packaging, medical devices and appliances, films for packaging, bottles and cups for beverages, any of which are based on plastic resin compositions as described above.
- 30 A molded article is preferred. The molding is in particular performed by injection-, blow-, compression- or rotational-molding, by thermoforming or extrusion.

A further preferred embodiment of the present invention relates to a monoaxially-oriented film or a biaxially-oriented film which has been formed by stretching a cast film based on resin compositions as described above.

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A multilayer system in which one or more layers contain a resin composition as described above is also of significance in the sense of the present invention.

The following example describes the present invention in more detail. All parts and percentages are given by weight unless indicated otherwise.

EXAMPLE:

Powdery propylene-ethylene copolymer (PP raco) with a melt flow index of 12 dg/min (measured at 230°C and 2.16 kg) is mixed with calcium stearate, IRGANOX B 215 (RTM) (= tris[2,4-di-tert-butylphenyl] phosphite and pentaerythrityl tetrakis[3-(3,5-di-tert-butyl-4-hydroxyphenyl)propionate] in a weight ratio of 2:1), Compound (I-2), Fischer—Tropsch wax (AdSperse 868 (RTM)) and Compound (II-1) in a Henschel mixer. Then, the formulations are compounded twice on a Berstorff ZE 25x46D (RTM) at 250°C. The granules obtained after compounding are used for injection molding on Arburg 320 S (RTM) at 240°C to produce 1 mm plaques. Haze, clarity and transmittance of the plaques are measured on a Haze-Gard plus (BYK Gardner (RTM)) according ASTM D1003 and the yellowness Index is measured on a datacolor machine according ASTM D1925.

	Formulation
PP raco	2.0 kg
Ca-stearate	750 ppm
Irganox B 215 (RTM)	1000 ppm
Compound (I-2)	2000 ppm
AdSperse 868 (RTM)	5000 ppm
Compound (II-1)	200 ppm
Haze	14.78
Yellowness Index	1.07

Compound (I-2):

Compound (II-1):

Claims:

1. An additive mixture containing the components (A), (B) and optionally (C), wherein component (A) is at least one compound of the formula (I)

$$(R)_{m} \longrightarrow (CHOH)_{p}$$

$$(I)$$

wherein

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p is zero or 1;

m and n are independently of one another an integer from zero to 3; and the radicals R are independently of one another C₁-C₈alkyl, C₁-C₈alkoxy, hydroxy, halogen, C₁-C₈alkylthio, C₁-C₈alkylsulfoxy or 2 radicals R form together with 2 adjacent carbon atoms of the unsaturated parent ring a 5- to 7-membered carbocyclic or heterocyclic ring; component (B) is at least one compound of the formula (II)

$$HO \xrightarrow{A_1} (CH_2) \xrightarrow{X} C - NH - NH - C - (CH_2) \xrightarrow{X} A_1$$

$$A_2 \xrightarrow{A_3} A_3 \xrightarrow{A_2} OH$$

$$(II)$$

wherein

15 x and y are independently of one another an integer from 2 to 10; the radicals A₁, A₂ and A₃ are independently of one another C₁-C₁₀alkyl, C₅-C₁₂cycloalkyl unsubstituted or substituted by 1 to 3 C₁-C₁₀alkyl; phenyl unsubstituted or substituted by 1 to 3 C₁-C₁₀alkyl; or C₇-C₁₂phenylalkyl, and the radicals A₂ and A₃ are additionally hydrogen;

- with the proviso that at least one of the radicals A₁ and A₂ is branched C₃-C₁₀alkyl, C₅-C₁₂cycloalkyl unsubstituted or substituted by 1 to 3 C₁-C₁₀alkyl; phenyl unsubstituted or substituted by 1 to 3 C₁-C₁₀alkyl; or C₇-C₁₂phenylalkyl; and **component (C)** is a lubricant or a mixture of lubricants.
- 25 2. An additive mixture according to claim 1 wherein p is 1;

m and n are independently of one another zero, 1 or 2; and the radicals R are independently of one another C_1 - C_4 alkyl;

x and y are independently of one another an integer from 2 to 6; the radicals A_1 , A_2 and A_3 are independently of one another C_1 - C_5 alkyl, cyclohexyl unsubstituted or substituted by one methyl; phenyl unsubstituted or substituted by one methyl; or 2-phenylpropyl, and

5 A₃ is additionally hydrogen.

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3. An additive mixture according to claim 1 wherein component (A) is at least one compound of the formulae (I-1), (I-2) and (I-3), and

CHOH

$$CH_2OH$$
 CH_2OH
 CH_2OH
 CH_2OH
 CH_2OH
 CH_2OH
 CH_3
 CH

component (B) is the compound of the formula (II-1).

$$(H_{3}C)_{3}C \qquad O \qquad O \qquad (C(CH_{3})_{3}$$

$$HO \longrightarrow (CH_{2})_{2} \longrightarrow (CH_{2})_{2} \longrightarrow (CCH_{3})_{3}$$

$$(H_{3}C)_{3}C \qquad (II-1)$$

- 4. An additive mixture according to claim 1 wherein component (C) is at least one lubricant selected from the group consisting of synthetic or natural waxes and amides of fatty acids.
 - 5. An additive mixture according to claim 1 wherein

component (C) is at least one lubricant selected from the group consisting of Fischer-Tropsch wax, high-pressure polyethylene wax, Ziegler-Natta polyethylene wax, metallocene polyethylene wax, Ziegler-Natta polypropylene wax, natural waxes and amides of fatty acids.

6. An additive mixture according to claim 1 wherein
 component (A) is at least one compound of the formulae (I-1), (I-2) and (I-3);

10 component (B) is the compound of the formula (II-1); and

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$$(H_3C)_3C$$

$$HO \longrightarrow (CH_2)_2$$

$$(CH_2)_2$$

$$(CH_2)_2$$

$$(II-1)$$

$$(H_3C)_3C$$

component (C) is at least one lubricant selected from the group consisting of Fischer-Tropsch wax, high-pressure polyethylene wax, Ziegler-Natta polyethylene wax, metallocene polyethylene wax, Ziegler-Natta polypropylene wax and stearamide, erucamide and oleamide.

7. A composition containing the components (I) and (II) wherein component (I) is a polypropylene homopolymer, random copolymer, alternating or segmented copolymer, block copolymer or a blend of polypropylene with another synthetic polymer; and

component (II) is the additive mixture according to claim 1.

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- **8.** A composition according to claim 7 wherein component (I) is a polypropylene homopolymer.
- **9.** A composition according to claim 7 wherein component (I) is a polypropylene random copolymer, alternating or segmented copolymer or block copolymer, containing one or more comonomers selected from the group consisting of ethylene, C_4 - $C_{20}\alpha$ -olefin, vinylcyclohexane, vinylcyclohexane, C_4 - C_{20} alkanediene, C_5 - C_{12} cycloalkandiene and norbornene derivatives.
- **10.** A composition according to claim 7 wherein component (I) is a polypropylene copolymer, manufactured by copolymerisation of at least 75 % by weight of propylene with ethylene or another alpha-olefin comonomer, which is selected from linear or branched butene, linear or branched pentene, linear or branched hexene and linear or branched octene.
- 11. The use of the additive mixture according to claim 1 as clarifying agent for a polypropylene homopolymer, random copolymer, alternating or segmented copolymer, block copolymer or a blend of polypropylene with another synthetic polymer.
- 12. A method for clarifying a polypropylene homopolymer, random copolymer, alternating or segmented copolymer, block copolymer or a blend of polypropylene with another synthetic polymer, which comprises incorporating therein an additive mixture according to claim 1.
- 25 **13.** An additive mixture containing the components (A') and (C'), wherein **component (A')** is at least one of the compounds of the formulae (I-1) and (I-2); and

$$H_3C$$
 O O CH_3 CH_3 CH_3 CH_3 CH_3

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component (C') is at least one lubricant selected from the group consisting of a synthetic wax, stearamide, erucamide and oleamide.

- 5 —14. A composition containing the components (I') and (II') wherein component (I') is a polypropylene homopolymer, random copolymer, alternating or segmented copolymer, block copolymer or a blend of polypropylene with another synthetic polymer; and component (II') is the additive mixture according to claim 13.
 - **15.** A method for clarifying a polypropylene homopolymer, random copolymer, alternating or segmented copolymer, block copolymer or a blend of polypropylene with another synthetic polymer, which comprises incorporating therein an additive mixture according to claim 13.
- 15 16. An additive mixture containing the components (A") and (C") wherein component (A") is the compound of the formula (I-3); and

$$H_3C$$
 O
 CH_3
 CH_3
 CH_2OH
 CH_3
 CH_3

component (C") is selected from the group consisting of stearamide, erucamide and oleamide.

- 17. A composition containing the components (I") and (II") wherein component (I") is a polypropylene homopolymer, random copolymer, alternating or segmented copolymer, block copolymer or a blend of polypropylene with another synthetic polymer; and
- 25 component (II") is the additive mixture according to claim 16.

- **18.** A method for clarifying a polypropylene homopolymer, random copolymer, alternating or segmented copolymer, block copolymer or a blend of polypropylene with another synthetic polymer, which comprises incorporating therein an additive mixture according to claim 16.
- 19. A composition containing the components (I"), (A") and (C") wherein component (I") is a polypropylene homopolymer or a polypropylene copolymer, manufactured by copolymerisation of at least 75 % by weight of propylene with ethylene or another alpha-olefin comonomer, which is selected from linear or branched butene, linear or branched pentene, linear or branched hexene and linear or branched octene;
 component (A") is the compound of the formula (I-3); and

$$H_3C$$
 O
 CH_3
 $CH_$

component (C"") is selected from the group consisting of Fischer-Tropsch wax, high-pressure polyethylene wax, Ziegler-Natta polyethylene wax, metallocene polyethylene wax, Ziegler-Natta polypropylene wax and stearamide, erucamide and oleamide.

Abstract

An additive mixture containing the components (A), (B) and optionally (C), wherein component (A) is at least one compound of the formula (I)

$$(R)_{m}$$
 $(CHOH)_{p}$
 $(CHOH)_{p}$
 $(R)_{n}$
 $(R)_{n}$

wherein

p is zero or 1;

m and n are independently of one another an integer from zero to 3; and the radicals R are independently of one another C₁-C₈alkyl, C₁-C₈alkoxy, hydroxy, halogen, C₁-C₈alkylthio, C₁-C₈alkylsulfoxy or 2 radicals R form together with 2 adjacent carbon atoms of the unsaturated parent ring a 5- to 7-membered carbocyclic or heterocyclic ring; component (B) is at least one compound of the formula (II)

$$HO \xrightarrow{A_1} (CH_2)_{\overline{X}} C - NH - NH - C - (CH_2)_{\overline{Y}} \xrightarrow{A_1} OH$$

$$(II)$$

wherein

x and y are independently of one another an integer from 2 to 10;

the radicals A_1 , A_2 and A_3 are independently of one another C_1 - C_{10} alkyl, C_5 - C_{12} cycloalkyl unsubstituted or substituted by 1 to 3 C_1 - C_{10} alkyl; phenyl unsubstituted or substituted by 1 to 3 C_1 - C_{10} alkyl; or C_7 - C_{12} phenylalkyl, and

the radicals A2 and A3 are additionally hydrogen;

with the proviso that at least one of the radicals A_1 and A_2 is branched C_3 - C_{10} alkyl, C_5 - C_{12} cycloalkyl unsubstituted or substituted by 1 to 3 C_1 - C_{10} alkyl; phenyl unsubstituted or substituted by 1 to 3 C_1 - C_{10} alkyl; or C_7 - C_{12} phenylalkyl; and **component (C)** is a lubricant or a mixture of lubricants.

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